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(54) STABILIZED TRANSPARENT RECEPTOR SHEET

(71) We, MINNESOTA MINING AND MANUFACTURING COMPANY, a corporation organized and existing under the laws of the State of Delaware, United States of America, of 3M Center, Saint Paul, Minnesota 55101, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to transparent receptor sheets. Particularly, the invention relates to the stabilization of receptor sheets capable of being imaged containing reducible metal soap and organic reducing agent.

In United States Patent No. 3,935,012, there are described light-sensitive intermediate sheets comprising (a) photolyzable heat-stable source of hydrogen halide, (b) acid-cleavable adduct of a first reactant and an organic compound containing a vinyl ether linkage, the adduct being capable of supplying a reducing agent when cleaved with acid, and (c) acid acceptor for the hydrogen halide in an amount less than sufficient to retain all of the hydrogen halide available from the photolyzable source upon exposure to a light image. When this sheet is exposed to a light image hydrogen halide is formed which cleaves the adduct so as to free a reducing agent at light-exposed areas. The intermediate sheet is then placed against a receptor sheet containing a second reactant (e.g., reducible metal soap and organic reducing agent such as sterically hindered phenol) and heated so as to induce a visible image-forming reaction. An image is formed in the receptor sheet due to reduction of a portion of the metal soap (to the metal) by reaction thereof with the liberated reducing agent from the light-struck areas of the intermediate sheet. This then catalyzes the heat-induced reduction of the remaining metal soap by the less reactive organic reducing agent originally present in the receptor to form a visible image.

Although the receptor sheet which has been imaged in accordance with the foregoing procedure is of good quality and is very easy to use, it has been found that those receptor sheets which include reducible metal soap and organic reducing agent tend to exhibit unacceptable increases in background optical density when exposed for a prolonged period of time (e.g. 2 hours or more) on a conventional overhead projector. During image formation, a small amount of material from the non-light-exposed areas of the intermediate sheet is transferred to the receptor sheet. Thus, when the imaged receptor sheet is used on an overhead projector, sufficient reducing agent is liberated so as to bring about the increase in background optical density. We have sought to overcome this problem of background optical density increase by including in the receptor sheet a small amount of aromatic dye.

Accordingly the present invention provides a receptor sheet for use in an image transfer process comprising a transparent support having at least one coating layer thereover including a film-forming binder, a reducible metal soap, an organic reducing agent and from 0.00007 to 0.005 parts by weight, per part of reducible metal soap, of a polycyclic aromatic dye compound exhibiting a development characteristic, as herein defined, of at least 150 seconds.

Optionally, the coating may also contain a toner (e.g. phthalazinone) for the metal image, or fillers or various other additives.

Preferably the support is a thin, flexible, transparent plastics film of a type which is known in the art as being useful for use in a projection transparency.

The improved receptor sheets of the present invention, after being imaged, may be projected for prolonged periods on conventional overhead projectors without objec-

tionable increase in the optical density of background areas. It has been suggested that the polycyclic aromatic dye stabilizes the receptor sheet by retarding the reduction of the metal soap by the liberated reducing agent in background areas.

The reducible metal soap present in the receptor sheet is preferably a silver soap of a long chain fatty acid (e.g. silver behenate, silver stearate), although other reducible metal soaps such as ferric stearate, gold stearate and cerium stearate are also useful. Other useful reducible metal soaps are known in the art.

The organic reducing agents which are useful in the receptor sheets are also well known in the art and include, as representative examples, pyrogaloll; 4 - azeloyl - bis - pyrogallol; 4 - stearoyl pyrogallol; gallacetophenone; ditertiarybutyl pyrogallol; gallic acid anilide; methyl gallate, ethyl gallate; normal and isopropyl gallate; butyl gallate; dodecyl gallate, gallic acid; ammonium gallate; ethyl protocatechuate; cetyl protocatechuate; 2,5-dihydroxy benzoic acid; 1-hydroxy-2-naphthoic acid; 2-hydroxy, 3-naphthoic acid; phloroglucinol; catechol; 2,3-naphthalene diol; 4-lauroyl catechol; sodium gallate; protocatechualdehyde; 4-methyl esculetin; 3,4-dihydroxy benzoic acid; 2,3-dihydroxy benzoic acid; hydroquinone; 4,4′-dihydroxy biphenyl; 3,4-dihydroxyphenylacetic acid; 4(3′,4′-dihydroxyphenylazo) benzoic acid; 2,2-methylene bis-3,4,5-trihydroxybenzoic acid; ortho- and paraphenylene diamine; tetramethyl benzidine; 4,4′,4″-diethylamino triphenylmethane; o-, m- and p-aminobenzoic acids; alpha and beta naphthols; 4-methoxy-1-naphthol, 1-hydroxy-dihydronaphthalene; and tetrahydroquinoline. These compounds are cyclic or aromatic compounds having an active hydrogen atom attached to an atom of carbon, oxygen or nitrogen which in turn is attached to an atom of the cyclic ring. They are capable of causing the reduction of metal ion and precipitation of metal on being dissolved at moderate temperature in a solution of aqueous metal salt in an organic solvent.

The reducible metal salt and organic reducing agent are preferably intermixed prior to coating, but may be applied in separate but contiguous layers if desired, with the aromatic dye being present in the layer containing the reducible metal salt.

Various types of film-forming binders may be used in the receptor sheets. For example, one may use cellulose propionate resin, polyethyl methacrylate resin, polyvinyl acetate, polyvinyl acetate/vinyl chloride copolymer, and cellulose acetate butyrate. Other conventional binders are well known in the art.

The relative amounts of reducible metal soap and organic reducing agent present in the receptor may vary, although there should be a sufficient amount of each present so as to be capable of forming a visible and distinct image when heated in the presence of the reactant (i.e., reducing agent) supplied by the intermediate sheet described above, Generally speaking, the amount of reducible metal soap present is from 4 to 12 parts by weight per part of organic reducing agent present.

The types of aromatic dyes which are useful in the receptor sheet to alleviate increase in background optical density have not been entirely definable in terms of chemical classification of dyestuffs. Fortunately, however, a simple test has been developed which accurately distinguishes between those aromatic dyes which are useful and those which are not. This test is used herein to define the development characteristics. This test procedure is as follows: A solution is first prepared using the following ingredients in the amounts shown:

Cellulose acetate propionate ("EAP 482—20"), available from Eastman)	5 grams
Methylethylketone	95 grams
Aromatic dye to be tested	0.10 millimoles
4-methoxy-1-naphthol	0.575 millimoles

Then 15 grams of this solution is poured into an aluminium dish (to a depth of 0.5 inch) and irradiated at a distance of 6 inches for 1 hour with a 150 watt General Electric tungsten filament light source. Thereafter the irradiated solution is coated (at 50 microns wet thickness) onto a receptor sheet (6 months or less in age and which is described below), followed by drying at 65°C. After drying, the receptor sheet is heated at 100°C. (212°F.). The time required for the optical density of the receptor sheet to show an increase to 0.85 is recorded as the "development characteristic" for the particular dye being tested. Those dyes which are useful in the present invention exhibit a "development characteristic" of at least 150 seconds. Preferably the dyes exhibit a "development characteristic" of at least 300 seconds. In this test procedure the optical density is measured using a conventional MacBeth Model TD 404A densitometer with a yellow filter.

The receptor sheet used in the foregoing test is prepared as follows: A silver

behenate dispersion is prepared using the following ingredients in the amounts shown:

	Parts by Wt.
Silver behenate	12.00
Tetrachlorophthalic anhydride	0.24
Stannous stearate	0.0072
Methylethylketone	87.7528
	100.0000

The above ingredients are mixed for one-half hour, then homogenized at 8000 psi, cooled to 80°F., and homogenized again at 8000 psi.

A binder solution is prepared with the following ingredients:

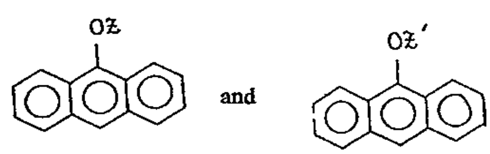
	Parts
Tenite H4® cellulose acetate butyrate resin (available from Eastman)	7.50
Elvacite 2042® polyethylmethacrylate resin (available from E. I. du Pont)	7.50
Methylethylketone	85.00
	100.00

A coating composition is then prepared using the following ingredients:

	Parts
Silver behenate dispersion	54.5531
Binder solution	43.6400
Phthalazinone	0.9819
2,6-di-t-butyl-p-cresol	0.7850
4,4'-methylene-bis-2,6-di-t-butylphenol	0.0400
	100.0000

The above ingredients are thoroughly blended and then coated onto a transparent plastics film at an orifice of 3.5 mils (87.5 microns) and dried in a forced air oven (3 minutes at 180°F.) to leave a dried, clear and transparent coating of 0.70 grams per square foot. A top coating is then applied using a 5% solution of cellulose acetate propionate in methylethylketone (coated at a wet thickness of 50 microns and dried to leave a dry coating having a thickness of 5 microns).

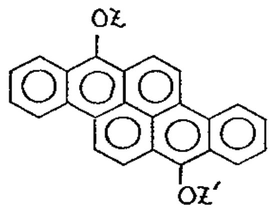
A preferred class of useful polycyclic aromatic dyes having a "development characteristic" of at least 150 seconds are oxygen-containing dyes comprising at least two moieties in conjugate relationship as part of a single chromophore, each moiety comprising three linearly kata condensed six-membered aromatic rings, an —OZ group being attached to the meso position of each moiety, wherein Z is a stable, monovalent radical; the —OZ group being a solubilizing group for the compound. Preferably, at least one auxochromic group comprising an atom having an atomic weight of at least 31 is bonded to the chromophore, the atom being attached directly to the chromophore. The above-mentioned moieties comprising three linearly kata condensed six-membered aromatic rings may be described pictorially as follows:



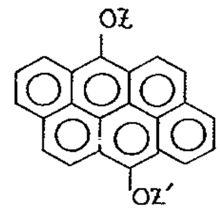
The aromatic rings may be homocyclic (carbon atoms) or heterocyclic, the hetero atoms being generally nitrogen. Of course, a single compound can contain both homocyclic and heterocyclic moieties of the type just described, or the compound may contain only homocyclic or only heterocyclic moieties.

These moieties are in conjugate relationship in the dye compound so that these moieties are part of a single chromophore (i.e., part of the same chromophore). A

chromophore may be defined as a group of atoms or electrons in a molecule which is chiefly responsible for an absorption band, as defined in Theory and Application of Ultraviolet Spectroscopy; Jaffe and Orchin; John Wiley & Sons, Inc., (1962). The two moieties may be bonded or condensed together in peri fashion, e.g.,



or



wherein the two moieties share atoms, or the moieties may be attached to each other in conjugate relationship through at least one linking moiety. The linking moieties are selected from (a) atoms which are at least trivalent and which are capable of forming covalent bonds, e.g., nitrogen, carbon, sulphur and oxygen, (b) ligands, having two or more atoms, which are at least bidentate (e.g., such ligands can be bidentate, tridentate, quadradentate, hexadentate, octadentate, etc.), and (c) covalent single or double bonds.

The size, chemical nature or structure of the linking moiety is not critical so long as the two moieties comprising the kata condensed aromatic rings are attached to each other in conjugate relationship such that they are part of a single chromophore.

The linking moiety, of course, can be a polycyclic structure (homocyclic or heterocyclic). Hetero atoms in the heterocyclic linking moieties are generally nitrogen, oxygen and sulphur.

In many of the compounds there are two linking moieties. For example, there may be two ligands, one ligand and a covalent bond or two covalent bonds as linking moieties in a single compound.

Preferably, these dye compounds have at least one auxochromic group bonded to the chromophore thereof. These preferred dyes are described in U.S. 3,819,664. The auxochromic groups may consist of one atom or of many atoms, so long as a heavy atom (i.e., having an atomic weight of at least 31), present as part of the auxochromic group, is bonded directly to the chromophore portion of the dye. An auxochromic group may be defined as a group bonded to a chromophore which influences the nature of the excited states, as defined in Theory and Application of Ultraviolet Spectroscopy; Jaffe and Orchin; John Wiley & Sons, Inc. (1962).

Preferred single atom auxochromic groups include chlorine, bromine, mercury, sulphur, iodine and selenium. Other useful single atom auxochromic groups include phosphorus (treated herein as having an atomic weight of 31), arsenic, tellurium, germanium, tin, lead and antimony. The auxochromic group may consist of more than one atom so long as a heavy atom present as part of the auxochromic group is directly bonded to the chromophore portion of the dye.

Although the —OZ and —OZ′ groups which are present on the dye compounds are also auxochromic groups, it has been found that the —OZ and —OZ′ groups are primarily solubilizing groups for the compound, i.e., they primarily determine the solubility of the dye compound in various solvent media into which the dye may be placed. Thus, the chemical structure and nature of the Z and Z′ radicals are not critical.

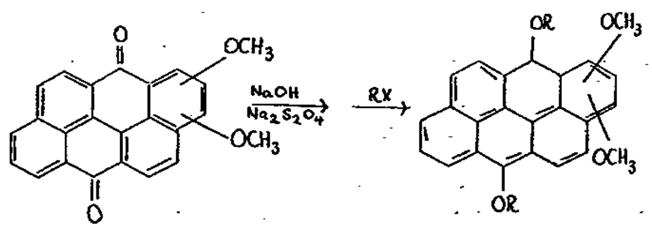
Generally, it may be said that Z and Z′ are monovalent radicals which are stable under ambient conditions and which do not cause decomposition of the chromophore portion of the dye compound. That is, these radicals do not oxidize or reduce the chromophore portion of the dye compound nor do they destroy or adversely affect the effectiveness of the compound as a dye. Within these limitations the —OZ and —OZ′ radicals can be stable derivatives of an inorganic acid, e.g., $—OSO_3Y+$ where Y is an alkali metal, alkaline earth metal, or ammonium ion; $—OPO_3R_2$ where R is hydrogen or a stable organic radical; and $—OPO_2R_2$ where R is hydrogen or a stable organic radical.

Insofar as organic radicals are concerned, Z and Z′ may be alkyl, cycloalkyl, substituted alkyl and cycloalkyl, alkenyl, alkynyl, aryl, polycyclic, acyl, alkaryl or aralkyl. Z and Z′ may be the same or different. Alkyl radicals having one carbon or

more are common Z radicals, and lower alkyl radicals containing from 1 to 6 carbon atoms are preferred, although long chain alkyls are also useful. Substituted alkyl radicals are herein defined to include alkyl radicals which are substituted with any moiety or group other than hydrogen atoms and other alkyl radicals.

In addition to the —OZ and —OZ′ solubilizing groups and the auxochromic groups the above classes of dye compounds may also be substituted with various other groups (e.g., fluorine, nitrile, hydroxy, alkyl, aryl, polycyclic, acyl, alkoxy) which are stable and do not cause decomposition of the chromophore portion of the dye compound.

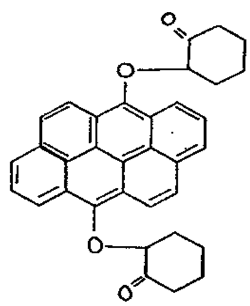
The dye compounds used in this invention can be prepared by independent synthesis but they are more conveniently prepared from precursor dyes. For example, Vat Violet 7 (Colour Index No. 59321) may be alkylated according to the following scheme with conventional alkylating agents:



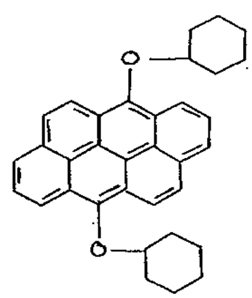
where RX represents agents such as alkyl iodide, alkyl tosylate, alkyl benzene sulphonate, and dialkyl sulphate. Other representative useful alkylated vat dyes include ethylated Vat Blue 18 (Colour Index No. 59815), and ethylated Vat Green 1 (Colour Index No. 59825).

To obtain compounds having R groups which are allyl, substituted allyl, propargyl, or substituted propargyl, alkylating agents such as allyl bromide and propargyl bromide may be used in the above reaction scheme.

To obtain compounds wherein R and R′ represent cycloalkyl radicals the dianion may be reacted with acrivated cycloalkyl halides, such as 2-chlorocyclohexanone, using the above reaction scheme. The resulting product has the following formula:



This product may be subsequently reduced via a Wolff-Kischner type reaction to yield a compound of the following formula:



To form an acyl derivative the vat dye is first reduced in the presence of zinc metal and an acid (e.g., acetic acid) followed by reaction of the reduced compound with an anhydride (e.g., acetic anhydride). Various other of such dye compounds can also be prepared using techniques well known in the art.

Other useful aromatic dyes not having the foregoing chemical make-up include, for example, octaphenyltetrazaporphyrin and erythrosin, which also exhibit a "development characteristic" of at least 150 seconds.

The amount of polycyclic aromatic dye present in the receptor sheet may be defined as an effective amount (i.e., an amount sufficient to reduce the increase in background optical density upon prolonged exposure on an overhead projector). Generally speaking, the tendency of the background optical density to increase is inversely proportional to the amount of aromatic dye present in the receptor. Thus, even minute amounts of dye present provide some usefulness. The maximum amount of dye present is determined either by the solubility limit of the dye in the binder or by the presence of an undesirable coloured tint in the sheet due to the dye. Within these limits the amount of aromatic dye present is from 0.00007 to 0.005 parts by weight per part of metal soap present.

The invention is further illustrated in the following examples, wherein the term "parts" refers to parts by weight unless otherwise indicated.

Example 1

A suitable receptor sheet is prepared using silver behenate, a sterically hindered phenol reducing agent, and an alkylated vat dye. The silver behenate dispersion was prepared using the following ingredients in the amounts shown:

	Parts
Silver behenate	12.00
Tetrachlorophthalic anhydride (development modifier)	0.24
Stannous stearate	0.0072
Methylethylketone	87.7528
	100.0000

The above ingredients were mixed for one-half hour, then homogenized at 8000 psi. cooled to 80°F., and homogenized again at 8000 psi.

A binder solution was prepared with the following ingredients:

	Parts
"Cellit PR-700" cellulose propionate resin (available from Bayer)	7.50
"Elvacite 2042" polyethylmethacrylate resin (available from E. I. du Pont)	7.50
Methylethylketone	85.00
	100.00

A coating composition is then prepared using the following ingredients:

	Parts
Silver behenate dispersion	54.5851
Binder solution	43.6400
Phthalazinone (toner for silver image)	0.9819
2,6-di-t-butyl-p-cresol	0.7850
Butylated Vat Violet 7 (Colour Index No. 59321) dye	0.0080
	100.0000

The above ingredients were thoroughly blended and then coated onto a transparent plastics film at an orifice of 6 mils (150 microns) and dried in a forced air oven (3 minutes at 180°F.) to leave a dried, clear and transparent coating of 0.0013 grams per square centimetre.

A top coating was then applied using a 5% solution of cellulose acetate propionate

in methylethylketone (coated at a wet thickness of 50 microns and dried to leave a dry coating having a thickness of 5 microns).

Example 2

An intermediate sheet was prepared using a coating composition comprising:

	Parts
Cellulose acetate butyrate	7.887
Solvent (methylethylketone)	91.00
Adduct of 4-methoxy-1-naphthol and dihydropyran (equimolar)	0.717
Tris-(tribromomethyl)-s-triazine	0.58
Spectral sensitizer (perylene)	0.052
Acid acceptor (diphenylguanidine)	0.016

The above composition was coated onto a clear polyester film from a 3 mil (75 microns) coating orifice and dried, all operations being conducted under appropriate safe light, to provide a photosensitive intermediate sheet.

The coated surface of the resulting intermediate sheet was then placed in contact with a printed original having light-absorptive image areas on a reflective white background, followed by uniformly exposing through the intermediate (i.e., in reflex position) to intense illumination from a bank of tungsten filament lamps for a time just sufficient to develop the coating completely at the background areas. A typical exposure time is 12 to 15 seconds. The image areas were also affected but to a significantly lesser extent due to absorption in the image areas of the original of a portion of the illumination. The exposed intermediate sheet was then placed with its coated surface against the coated face of the receptor sheet of Example 1 (e.g., between rolls or platens) and heated for 4 to 5 seconds at 125 to 140°C. A sharp negative copy of the graphic original was obtained on the receptor sheet.

The receptor sheet was then placed on a "Model AR-66" overhead projector (available from Minnesota Mining and Manufacturing Company) for 16 hours with no perceptible increase in background optical density.

This result was distinguished from that observed when the example was repeated using a receptor sheet having no polycyclic aromatic dye therein. In this latter case there was a 1000% increase in background optical density when left on the above projector for 16 hours.

Examples 3 to 7

Several receptor sheets were prepared, each having a different amount of polycyclic aromatic dye therein. A stock solution was first prepared using the following ingredients:

	Parts
Silver behenate dispersion from Example 1	54.585
Phthalazinone	0.982
Binder solution from Example 1	43.585
2,6-di-t-butyl-p-cresol (sterically hindered phenol reducing agent)	0.793

To similar samples of the above stock solution (each sample containing 25 grams of silver behenate soap) were added varying amounts of butylated Vat Violet 7 (polycyclic aromatic dye), as shown in Table I:

TABLE I

Example No.	Polycyclic Dye Added (Grams)
3 (Comparative)	0
4	0.002
5	0.004
6	0.008
7 (Comparative)	0.016

Each solution was then knife coated onto separate samples of 2 mil (50 microns) thick polyester film using a 100 micron orifice. The samples were then air dried at 190°F. (88°C.) to leave a dried coating 20 microns thick, after which a protective top coating of cellulose acetate butyrate was applied (5 microns, dry thickness).

The resulting receptor sheets were then separately heated against previously exposed photosensitive intermediate sheets of the type described in Example 2. The so-imaged receptor sheets were then each placed on a "Model AR-66" overhead projector and the background optical density of each sample after 4, 12 and 30 hours was compared to the initial background optical density. The optical density was measured using a MacBeth densitometer (using a gold filter). The results are shown in Table II below.

TABLE II

	% Background Optical Density Increase		
Example No.	4 hours	12 hours	30 hours
3	200	500	1000
4	130	150	220
5	85	140	230
6	50	75	100
7	0	0	15

Example 8

The receptor sheet of Example 1 was prepared except that the silver behenate was replaced with an equal amount by weight of ferric stearate. The resulting receptor sheet was imaged using the intermediate sheet and technique of Example 2. A blue image on a buff background was obtained. Upon overhead projection the background optical density remained stable for a prolonged period.

Example 9

A receptor sheet was prepared and imaged according to Example 8 except that the ferric stearate was replaced with an equal amount by weight of gold stearate. A purplish-black image on a pale purple background was obtained. Upon prolonged overhead projection the background optical density remained stable.

Example 10

A receptor sheet was prepared and imaged according to Example 8 except that the ferric stearate was replaced with an equal amount by weight of cerium stearate. A blue image on a colorless background was obtained. Upon prolonged overhead projection the background optical density remained stable.

Example 11

A receptor sheet was prepared and imaged according to Example 1 except that the 2,5-di-t-butyl-p-cresol was replaced with 0.785 parts by weight of 4,4'-methylene-bis-2,6-di-t-butylphenol. Upon imaging of the receptor using the technique of Example 2 a dense black image was obtained. Upon prolonged overhead projection the background optical density remained stable.

When this example was repeated, but without the presence of butylated Vat Violet 7 (Colour Index No. 59321) in the receptor sheet, the background optical density increased approximately ten-fold after 12 hours of overhead projection.

Examples 12 to 13

Two separate receptor sheets were prepared using the procedure of Example 1, except that one sample (Example 12) contained no polycyclic aromatic dye and the other sample (Example 13) contained ethylated Vat Blue 18 in place of butylated Vat Violet 7. Upon imaging in accordance with Example 2 a dense black image was obtained. Upon overhead projection the background optical density at various times was measured. The results are shown in Table III.

TABLE III

	Background Optical Density		
Example No.	Initial	4 Hours	12 Hours
12	0.10	0.16	0.54
13	0.07	0.08	0.11

Examples 14 to 15

Two separate receptor sheets were prepared using the procedure of Example 1, except that one sample (Example 14) contained no polycyclic aromatic dye and the other sample (Example 15) contained octaphenyltetrazaporphyrin in place of butylated Vat Violet 7. After imaging as in Example 2, the receptor was placed on an overhead

projector for a prolonged period and the background optical density was measured at various times. The results are shown in Table IV.

TABLE IV

	Background Optical Density		
Example No.	Initial	4 Hours	12 Hours
14	0.06	0.14	1.15
15	0.06	0.07	0.09

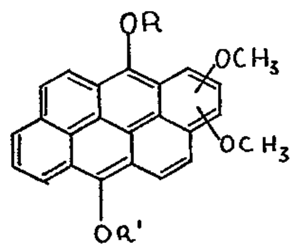
Other variants are possible within the scope of the present invention.

WHAT WE CLAIM IS:—

1. A receptor sheet for use in an image transfer process comprising a transparent support having at least one coating layer thereover including a film-forming binder, a reducible metal soap, an organic reducing agent and from 0.00007 to 0.005 parts by weight, per part of reducible metal soap of a polycyclic aromatic dye compound exhibiting a development characteristic, as herein defined, of at least 150 seconds.

2. A receptor sheet as claimed in Claim 1, wherein the dye compound comprises at least two moieties in conjugate relationship as part of a single chromophore, each moiety comprising three linearly kata condensed six-membered aromatic rings having an —OZ group attached to the meso position of each moiety, wherein Z is a stable, monovalent radical and wherein the —OZ group is a solubilizing group for the dye compound.

3. A receptor sheet as claimed in Claim 1 or 2, wherein the dye compound is of the formula



where R and R′ are each an alkyl group containing from 1 to 6 carbon atoms.

4. A receptor sheet as claimed in Claim 3, wherein R is a butyl group.

5. A receptor as claimed in claim , wherein the dye compound is octaphenyltetrazaporphyrin or erythrosin.

6. A receptor sheet as claimed in any of the preceding claims, wherein the reducible metal soap is silver behenate, silver stearate, ferric stearate, gold stearate or cerium stearate.

7. A receptor sheet as claimed in any one of the preceding claims, wherein the organic reducing agent is a sterically hindered phenol.

8. A receptor sheet as claimed in Claim 7, wherein the organic reducing agent is 2,6-di-t-butyl-p-cresol, or 4,4′-methylene-bis-2,6-di-t-butylphenol.

9. A receptor sheet as claimed in any one of Claims 1 to 6, wherein the organic reducing agent is 4-methoxy-1-naphthol.

10. A receptor sheet as claimed in any one of the preceding claims, wherein the dye compound is ethylated Vat Blue 18 (Colour Index No. 59815).

11. A receptor sheet in accordance with Claim 1 and substantially as herein described with reference to the Examples.

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